



IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re Application of:

Masahide Ishikawa

Application No. 10/500,867

Filed: July 21, 2004

For: AN AGENT FOR SUPPRESSING  
TRANSFER OF ODOR AND TASTE  
ORIGINATING FROM A  
DIACETAL, A DIACETAL  
COMPOSITION COMPRISING THE  
AGENT FOR SUPPRESSING  
TRANSFER OF ODOR AND TASTE,  
A POLYOLEFIN NUCLEATING  
AGENT COMPRISING THE  
COMPOSITION, A POLYOLEFIN  
RESIN COMPOSITION AND A  
MOLDED PRODUCT COMPRISING  
THE NUCLEATING AGENT

)  
) Group Art Unit: 1796  
)  
) Examiner: Nerangis, Vickie Marie  
)  
) Confirmation No.: 8698  
)

Commissioner for Patents  
PO Box 1450  
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Sir:

**DECLARATION UNDER 37 C.F.R. § 1.132**

I, Reira Ikoma, do hereby make the following declaration:

1. I am a Japanese citizen, residing at 1-250-1-202, Kyomachi, Fushimi-ku, Kyoto-shi, Kyoto 612-8083, Japan.
2. I graduated from Wakayama National College of Technology, Department of Materials Science in March, 2005.
3. I began my employment with NEW JAPAN CHEMICAL CO., LTD., the assignee of the above-identified application, in April, 2005. Since April, 2005, I have been engaged in the research and development of additives for polymer.

4. I am familiar with the subject matter of said application as well as the disclosures in the cited references.

5. I verify that a resin composition containing the following combinations of components (B) and (C) for use with diacetal (A) in an agent for suppressing the transfer of odor and taste originating from diacetal is excellent in suppressing aldehyde generation, odor, and the transfer of taste:

a combination of (B1) and (C1), (C3), or (C4)

a combination of (B1) and (C3) or (C4).

Component (B):

(B1) saturated or unsaturated aliphatic alcohol

(B2) saturated or unsaturated aliphatic carboxylic acid having at least one hydroxyl group

Component (C):

(C1) • saturated or unsaturated aliphatic alcohol sulfuric ester salt

• polyoxyethylene alkyl or alkenyl ether sulfuric ester salt

• polyoxyethylene alkyl phenyl ether sulfuric ester salt

• sulfuric ester salt of polyhydric alcohol fatty acid partial ester formed from polyhydric alcohol and saturated or unsaturated fatty acid

• saturated or unsaturated fatty acid monoalkanol amide sulfuric ester salt

(C3) • dialkanolamine

• trialkanolamine

• di(alkyl or alkenyl) methylamine

(C4) • a mixture of (C1) and (C3)

Experimental Example 1 and 2 (corresponding to Examples 26 and 27 of the description in  
USSN 10/500,867)

1,3:2,4-di(p-methylbenzylidene)sorbitol (hereinafter referred to as "Me-DBS"), 12-

hydroxystaric acid shown in Table I, and an amine shown in Table I were stirred for one hour in methanol (weighing 6 times as much as Me-DBS) under reflux to give a mixture in a white paste form. Then, the methanol was removed under reduced pressure, and the result was dried in a vacuum drier for one hour under the pressure of 133 Pa at a temperature of 80°C, to obtain a powdery sample (hereinafter referred to as "Me-DBS composition").

100 weight parts of an isotactic random polypropylene resin with an ethylene content of 3.0 wt% (MFR = 20 g/10 minutes; hereinafter referred to as "r-PP") was blended with a 0.2 weight part of the Me-DBS composition, a 0.05 weight part of tetrakis[methylene-3-(3,5-di-tert-butyl-4-hydroxyphenyl)propionate]methane (trade name "Irganox 1010", made by Ciba Specialty Chemicals) and a 0.05 weight part of calcium stearate, and these components were mixed in a Henschel mixer. Then the mixture was melt kneaded using a single screw extruder having a diameter of 25 mm at a resin temperature of 240°C and pelletized. The odor evaluation of the obtained pellets was carried out by the following dry method. Table I shows the results.

#### Odor evaluation of pellets by dry method

A 225 ml glass bottle in which 60 g of pellets were sealed was left to stand in a constant temperature bath of 80°C for 2 hours, and cooled to room temperature. Immediately after that, the odor intensity was rated by ten panelists. The rating criteria used by the panelists were as follows: zero point, no unpleasant odor; 1 point, slightly unpleasant odor; 2 point, distinct unpleasant odor; 3 point, strong unpleasant odor. Total value of the points determined by the ten panelists was used for evaluation.

The injection molding of the obtained pellet was carried out at a resin temperature of 260°C and a mold temperature of 40°C to give a test piece. Odor evaluation by dry and wet methods, measurement of aldehyde generation and taste evaluation was performed by the following methods using the obtained injection-molded product. In addition, measurement of crystallization temperature (Tc) and a haze value (%) was performed for evaluating the nucleating agent performance.

#### Method of measuring crystallization temperature (Tc)

According to JIS K 7121, the crystallization temperature was measured using a differential scanning calorimeter trade name "DSC7" from Perkin Elmer). The higher the Tc value, the quicker the crystallization rate is, and the molding cycle can be shortened.

#### Haze value (improvement in clarity)

The haze value was measured using a haze meter from Toyo Seki Seisakusho according to JIS K 6714 and JIS K 6717. The smaller the measured value, the better the clarity is.

Subsequently, the obtained injection-molded product was subjected to the odor evaluation by the following dry and wet methods.

#### Odor evaluation of injection-molded product by dry method

A 225 ml glass bottle in which 20 g of test piece were sealed was left to stand in a constant temperature bath of 80°C for 2 hours, and cooled to room temperature. Immediately after that, the odor intensity was rated by ten panelists. The rating criteria used by the panelists were as follows: zero point, no unpleasant odor; 1 point, slightly unpleasant odor; 2 points, distinct unpleasant odor; 3 points, strong unpleasant odor. Total value of the points determined by the ten panelists was used for evaluation.

After sealing 20 g of test piece and 140 g of deionized water in a 225 ml glass bottle, the bottle was left in a constant temperature bath of 80°C for 2 hours and cooled to room temperature. Odor evaluations, measurement of aldehyde generation and taste evaluation were performed by the following methods using the obtained solution as an evaluation sample.

#### Odor evaluation of infection-molded product by wet method

The evaluation sample was subjected to the odor intensity rating by ten panelists. The rating criteria used by the panelists were as follows: zero point, no unpleasant odor; 1 point, slightly unpleasant odor; 2 points, distinct unpleasant odor; 3 points, strong unpleasant odor. Total value of the points determined by the ten panelists was used for evaluation.

#### Aldehyde generation

Aldehyde content of the evaluation sample was measured using a high performance liquid chromatography. The aldehyde amount was expressed in  $\mu\text{g}/\text{PPg}$ , i.e.  $\mu\text{g}$  per 1 g of test

piece.

#### Taste evaluation of injection-molded product

Ten gram of the evaluation sample was placed in a cup made of glass and subjected to the taste evaluation by ten panelists. In the taste evaluation, ten panelists evaluated the evaluation sample in comparison with control samples prepared by the following method, wherein the rating criteria used by the panelists were as follows: zero point, no difference; 1 point, slight difference; 2 points, distinct difference. Total value of the points determined by the ten panelists was used for evaluation.

#### <Preparation of control samples for taste evaluation>

The solutions as control samples for taste evaluation were prepared in the same manner as in Example 1 of the description in USSN 10/500,867 except that DBSs were not added.

In the diacetal compositions shown in Table I and II, the amount of component (B) is expressed as a percentage (% by weight) based on the total amount of components (A) and (B). Accordingly, the percentage of component (A), MA (wt%), can be calculated as follows: MA=100-MB, wherein MB is the percentage of component (B) (wt%).

#### Experimental Examples a1 to a8

Pellets were obtained from mixtures of a resin, component (A), component (B), and component (C) in the same manner as in Experimental Example 1 except that the compounds shown in Table I were used.

Note that "h-PP" shown in Table I indicates isotactic homopolypropylene resin (MFR = 30 g/10 minutes); "DBS" indicates 1,3:2,4-dibenzylidene sorbitol); "Et-DBS" indicates 1,3:2,4-di(p-ethylbenzylidene)sorbitol; and "3,4-DMDBS" indicates 1,3:2,4-di(3,4-dimethylbenzylidene)sorbitol.

Each obtained pellet was evaluated in the same manner as in Experimental Example 1. Table I shows the results.

#### Comparative Experimental Examples 1 to 5 (corresponding to Examples 1 to 5 described in USSN 10/500,867)

Pellets were obtained from mixtures of a resin, component (A), and component (B) in the same manner as in Experimental Example 1 except that the compounds shown in Table II were used and component (C) was not used. Each obtained pellet was evaluated in the same manner as in Experimental Example 1. Table II shows the results.

Comparative Experimental Examples b1 to b9

Pellets were obtained from mixtures of a resin, component (A), and component (C) in the same manner as in Experimental Example 1 except that the compounds shown in Table II were used and component (B) was not used. Each obtained pellet was evaluated in the same manner as in Experimental Example 1. Table II shows the results.

Table I

Ex.	Resin	Diacetal composition			Crystallization temp. (°C)	Order evaluation		Aldehyde generation (μg/PPg)	Taste evaluation		
		Component (A)	Component (B)	Amount (wt.%)		Amount (wt.%)	Component (C)	Haze value (%)	Pellet	Injection product	
1	r-PP	Me-DBS	12-hydroxy-stearic acid	2.5	Diethanolamine	2.5	128	13	Dry method	Dry method	3.3
2	r-PP	Me-DBS	12-hydroxy-stearic acid lauryl alcohol	2.5	Trisopropanolamine	2.5	127	13	Dry method	Wet method	3.2
a1	r-PP	Me-DBS	lauryl alcohol	2.5	Na lauryl sulfate	2.5	127	12	Dry method	Dry method	2.6
a2	r-PP	Me-DBS	stearyl alcohol	2.5	Na lauryl sulfate	2.5	127	12	Dry method	Wet method	2.6
a3	r-PP	Me-DBS	stearyl alcohol	2.5	Na polyoxyethylene (3 moles added) lauryl ether sulfate	2.5	127	13	Dry method	Dry method	2.7
a4	r-PP	DBS	stearyl alcohol	2.5	Na lauryl sulfate	2.5	120	22	Dry method	Dry method	2.8
a5	r-PP	Et-DBS	stearyl alcohol	2.5	Na lauryl sulfate	2.5	125	18	Dry method	Dry method	2.6
a6	r-PP	3,4-DMDBS	stearyl alcohol	2.5	Na lauryl sulfate	2.5	128	12	Dry method	Dry method	2.2
a7	h-PP	Me-DBS	stearyl alcohol	2.5	Na lauryl sulfate	2.5	133	22	Dry method	Dry method	2.9
a8	r-PP	Me-DBS	stearyl alcohol	2.5	Trisopropanolamine	2.5	127	12	Dry method	Dry method	2.9

Table II

Com. Ex.	Resin	Diacetal composition			Crystalli- zation temp. (°C)	Haze value (%)	Order evaluation		Aldehyde generation (μg/PPg)	Taste evaluation	
		Component (A)	Component (B)	Amount (wt.%)			Component (C)	Amount (wt.%)	Dry method		
1	r-PP	Me-DBS	lauryl alcohol	2.5	-	-	127	12	8	10	9
2	r-PP	Me-DBS	myristyl alcohol	2.5	-	-	128	13	8	9	9
3	r-PP	Me-DBS	palmityl alcohol	2.5	-	-	127	12	8	9	9
4	r-PP	Me-DBS	stearyl alcohol	2.5	-	-	127	13	7	10	9
5	r-PP	Me-DBS	12-hydroxy- stearic acid	2.5	-	-	128	11	7	9	8
b1	r-PP	Me-DBS	-	-	Na lauryl sulfate	2.5	128	15	9	10	10
b2	r-PP	Me-DBS	-	-	Na lauryl sulfate	2.5	128	15	9	10	10
b3	r-PP	Me-DBS	-	-	Na polyoxyethylene (3 moles added) lauryl ether sulfate	2.5	127	14	10	9	9
b4	r-PP	DBS	-	-	Na lauryl sulfate	2.5	120	22	7	8	7
b5	r-PP	Et-DBS	-	-	Na lauryl sulfate	2.5	126	18	10	10	9
b6	r-PP	3,4-DMDBS	-	-	Na lauryl sulfate	2.5	128	12	9	9	8
b7	h-PP	Me-DBS	-	-	Na lauryl sulfate	2.5	133	21	8	10	10
b8	r-PP	Me-DBS	-	-	Triisopropanolamine	2.5	128	13	10	10	8
b9	r-PP	Me-DBS	-	-	Diethanolamine	2.5	128	14	9	10	7

## Results and Considerations

Table II shows that the amount of aldehyde generation in resin compositions that contain only component (B) or component (C) was extremely high, at 3.7 to 4.7 µg/PPg, i.e., aldehyde generation could not be sufficiently suppressed. In addition, pellets and injection-molded products of these compositions had unpleasant odors in the range of 7 to 10 in the odor evaluation. Further, the rating of the taste evaluation of the injection-molded products was also high, at 7 to 11.

In contrast, Table I shows that the amount of aldehyde generation was 2.2 to 3.3 µg/PPg in resin compositions in which a combination of specific compounds was used in the mixtures of components (A), (B), and (C), i.e., aldehyde generation was sufficiently suppressed. Accordingly, compared to Comparative Experimental Examples, pellets and injection-molded products of these compositions have reduced unpleasant odors in the range of 3 to 5 in the odor evaluation, and the rating of the taste evaluation of the injection-molded products was also low, at 2 to 5, in these compositions.

Based on the above, it was proven that a resin composition containing a combination of specific components (B) and (C) with diacetal (A) in an agent for suppressing the transfer of odor and taste originating from diacetal can suppress aldehyde generation and, as a result, can fully demonstrate the effects of suppressing odor and the transfer of taste.

6. I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under section 1001 of Title 18 of the United States Code, and that such willful false statements may jeopardize the validity of any patent issued on this application.

Date: October 20, 2009,

By: Reira Ikoma  
Reira Ikoma